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Title of Invention:

## **Integrated Processing of Biomass and Liquid Effluents**

### Related Applications:

Australian Patent Application No: 51888/00; Filing Date: August 8, 2000; Relationship: Equivalent Claims

US Provisional Patent Application No: 60/307,712; Filing Date: 07/26/01; Relationship: Equivalent Claims

### Background of Invention

This invention relates to a process for the manufacture of multiple valuable products from various wastes generated during the production and recovery of cane sugar, as well from other agricultural cellulosic biomass materials.

The cane sugar industry generates vast quantities of liquid wastes and solid residues during the production of its primary product. These include sugar cane bagasse and liquor streams containing large quantities of low-grade sugars. Bagasse is usually burned inefficiently in boilers to generate steam and power. The liquid waste streams, mostly molasses, are usually sold as low-grade cattle feed additives.

The cane sugar industry is normally a highly competitive, low profit industry. One approach to improve profitability is to convert these wastes and residues into saleable products that can add to the revenues of the industry. The value of this approach has been recognized for a number of years and considerable research and commercial development has been undertaken to identify useful products, such as furfural and papermaking pulp that can be generated from these wastes.

A new type of pulping technology, known as organosolv pulping, has distinct advantages for the cane sugar industry. It has almost no environmental problems, is less capital

intensive than kraft, it produces multiple co-products, is ideally suited to pulping non-wood biomass that contain high levels of inorganic materials, and can be profitably operated on a much smaller scale than conventional pulping processes. Organosolv pulping has been described in numerous patents and publications including *US Patent No. 3,585,104 Kleinert*, *US Patent No. 4,100,016 Diebold et al.*, *US Patent No. 4,496,426 Baumeister et al.* Although organosolv pulping has numerous advantages over conventional chemical pulping methods, such as the kraft, sulfite and the soda processes, in the pulping of wood and other woody biomass resources, it suffers from a disadvantage of significant losses of the relatively costly organic solvent from the process. If the solvent employed is ethanol the environmental consequences of this loss is minor, but the economic consequences can be important, since they can make the operating costs of an organosolv process higher than those for conventional chemical pulping processes. This requires the provision of substantial quantities of expensive make-up solvent to the process. This higher operating cost is one major reason that has held back the commercial acceptance of organosolv pulping processes.

One approach to reducing the cost of make-up solvent required by an organosolv pulp mill, to make it economically attractive, is to produce the required make-up solvent on site. For an ethanol-based organosolv pulp mill a fermentation facility can be constructed adjacent to the pulp mill for this purpose. However, if constructed on a small size sufficient for the make-up needs of a small pulp mill, the relative capital costs of such a fermentation facility would make it economically unattractive. Furthermore, if the pulp mill uses wood as a raw material, the fermentation plant would need to purchase fermentation feedstock, such as sugar or starch, at considerable cost. This problem can be overcome for an ethanol-based organosolv pulp mill processing agricultural residues, such as bagasse. Such a pulp mill processing bagasse would ideally be situated adjacent to a cane sugar mill, since the bagasse is a low value product of these mills. Therefore no transportation costs would be incurred for the raw material. Another low value by-product of the sugar mill is molasses, a high sugar content liquid by-product. It is well recognized that molasses

can be readily fermented commercially to yield ethanol. It therefore represents a low cost fermentation feedstock present at the sugar mill.

- 5                    Even more attractive for the cane sugar industry is that ethanol-based organosolv pulping is highly compatible with an agricultural economy. Its primary process chemical is ethanol, which can be easily produced by fermentation of waste sugars and starch. Furthermore, many of the co-products of the process
- 10 find immediate use and value in agriculture, such as animal feed supplements and slow release fertilizer and pesticides. This invention solves the problem of the high capital cost of a dedicated small fermentation plant to provide make-up ethanol by physically integrating the fermentation of molasses into the process
- 15 equipment of an organosolv pulp mill. A single mill consisting of organosolv pulping of biomass residues Integrated with the processing and fermentation of aqueous waste streams will result in major profits for the cane sugar industry. At the same time such a strategy will result in a high degree of environmental protection
- 20 and support for the development of adjacent industrial activities based on the co-products of this process.

### Brief Summary of the Invention

5 The present invention provides a process for delignifying biomass fibrous residues comprising digesting biomass fibrous residues in a mixture of ethanol and water in a digester at elevated temperature and pressure.

10 The process would include the step of treating spent liquor to recover lignin, acetic acid, furfural, xylose and other co-products, as well as recovering alcohol for re-use in the process and subjecting various liquid sugar waste streams of the process and from a cane sugar mill to fermentation in order to produce ethanol and other fermentation products which may be used in the process. The recovery of ethanol from the spent  
15 cooking liquors and the recovery of ethanol from the fermentation liquors would be accomplished in the same process equipment. These two processes, the organosolv pulping and the fermentation of waste sugar streams, would be accomplished in the same plant, which would allow equipment, heat and energy integration providing considerable economic advantages. This  
20 process concept is applicable to other biomass residues as well as cane-sugar residues. The potential advantages of integrating these two formerly separate activities into a single operating unit are numerous. They include lower total capital costs, combining different liquor streams for common product recovery, opportunities for process heat and energy reduction, internal process chemical production, reduced transportation costs, substantial environmental improvement and the potential for the use of larger, more efficient equipment in a shared operation.  
25 Separate facilities operating in isolation may not be economic. This identifies the unit processes and the product flows in an integrated total process that would provide these advantages to the cane sugar industry.

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The distillation tower in the process could recover not only ethanol for recycle, but also furfural and ethyl acetate, two valuable products that are generated during the bagasse cooking stage. Both can be sold, probably as crude products suitable for

upgrading at a centralized facility. Such activities would encourage the formation of additional local industries designed to support the sugar cane processors using the technology described in this invention. Other local industries could take the lignin produced in these mills and convert it to value-added products, such as concrete admixtures and dye dispersants. Other options presented by this invention include the recovery of xylose (a sugar present in large quantities, mostly as xylan, in bagasse). This could be sold in the world market for pure xylose that is used as a starting material in the production of the anti-caries sweetener, xylitol. Xylose can also be converted to furfural. If market prices support this option then xylose recovery could be maximized by extended steaming of the bagasse prior to cooking. Xylose would be recovered from the steaming condensate.

Utilizing this invention could lead to higher value pulps which would have high alpha-cellulose content and therefore be suitable for rayon production. The result for the cane sugar industry of practicing this invention would be the elimination of costly environmental control operations and the production of significant revenues from the sale of several value-added products. These products would in turn create opportunities for the introduction of ancillary industries.

## Description of the Drawing

In the accompanying drawing, which illustrates by way of example the embodiment of this invention, Fig.1 is a flow diagram identifying the unit process steps of an integrated process for the production of pulp and several useful by-products from bagasse, a biomass residue of the cane sugar industry. While not every unit process is essential for the economic success of the invention, the combination of all these unit processes provides maximum utility of the invention.

## Detailed Description of the Preferred Embodiment

The present invention is directed at a single integrated process that converts biomass residues from the cane sugar industry, sometimes referred to as bagasse, into a series of valuable products including, but not limited to, papermaking pulps. The process is integrated with the element of fermentation of waste and low-grade molasses to

5 This element utilizes some of the alcohol generated in the fermentation element. The organosolv element generates products such as lignin, xylose, furfural, acetic acid and pulp for use in papermaking, dietary fibre, or as chemical cellulose. Much utility is gained by integrating these several elements into a single process.

10 The advantages include heat and energy reduction through process integration, capital reduction through the co-processing of various process streams and waste minimization opportunities through the combining of several process streams. These elements and advantages are illustrated in the process flow diagram, Fig.1.

The process starts with the preparation of the bagasse into a form suitable for packing into a pressure vessel,<sup>1</sup> identified in Fig.1 as a digester. The preferred form is into stem sections of approximately ten centimeters in length, but any similar form is appropriate. A compressed pellet is also appropriate as feed for the digester. As illustrated the digester is one of a series of batch digesters that may be rotating spheres, or a continuous conveyed inclined or horizontal tube configuration, but could also be a vertical tubular batch or continuous design. In Fig.1 a preferred configuration of a rotating globe batch digester configuration is illustrated. The operations described below for one digester are identical to those for the additional digesters that are operated sequentially at appropriate intervals to allow optimal use of the remaining equipment in the process.

Following preparation of the fibrous residue (bagasse) into the useful form described above it is conveyed by conveyer, **41**, to the top of the digester for loading into the digester. Once the digester, **1**, is filled with a pre-determined amount of bagasse, the conveyer is stopped and the digester is closed. An exhaust valve, **46**, located behind a screen, **42**, in the bottom of the digester is now opened and low-pressure steam (less than 50 psi) is allowed to enter the top of the digester through a pipe, **45**. This steaming, which is required to displace air in the fibre bed, continues until temperature sensors in the exhaust line indicate that steam is exiting from the bottom of the digester. Any condensate of the steaming exits through

the same line. Alternatively, if the moisture content of the bagasse is too high, nitrogen gas may be substituted for steam for the air displacement. All valves are now closed and the pump, **43**, in the line exiting the 2<sup>nd</sup> liquor tank is turned on. The 2<sup>nd</sup> liquor tank is full of aqueous alcohol at the desired concentration and temperature. This liquor was used as a wash liquor from a previous digester cook and was retained between cooks in the 2<sup>nd</sup> Liquor Tank, **3**. The preferred alcohol concentration in water is in the range of 35%-70% (w/w) and the preferred temperature is in the range of 170°-205°C. This liquor is pumped through a heat exchanger, **44**, to maintain its desired temperature and then into the top of the digester through the top liquor line. Once the digester starts to fill, liquor exits the bottom of the digester from behind a screen, **42**, constructed around the outlet line at the bottom of the digester, from where it is returned to the top of the 2<sup>nd</sup> Liquor Tank, **3**. This hot liquor circulation is continued for the appropriate time necessary to raise the contents of the digester to the desired cooking temperature. At this point the liquor exit valve, **46**, is closed and the desired weight of hot liquor, usually 2 to 5 times the dry weight of the bagasse, is pumped into the digester from the 2<sup>nd</sup> Liquor Tank. Liquor flow is then stopped, steam is continually sent to a jacket surrounding the digester to maintain its temperature at the desired cooking temperature and the globe digester is rotated for the desired cooking time. This time is normally between 30 minutes and 3 hours, with the preferred time being between one and two hours. At the end of this time the rotation of the digester is stopped with the liquor outlet line and surrounding screen at the bottom of the digester. Part of the hot black liquor is then flashed into a Flash Tank, **6**. The valve at the top of the 2<sup>nd</sup> Liquor Tank is then closed and the return liquor is diverted to the Spent Liquor Tank. Residual liquor in the 2<sup>nd</sup> Liquor Tank is pumped down to a level sufficient to keep the suction side of the pump flooded. Liquor remaining in the Digester is drained through the lower screens into a drain line from where it is also pumped to the Spent Liquor Tank. Next, clean aqueous alcohol at the concentration and temperature previously described is pumped from the 1<sup>st</sup> Liquor Tank, **2**, into the top and bottom lines of the Digester and returned to the 2<sup>nd</sup> Liquor Tank, **3**, using the appropriate valves and pumps. After the 1<sup>st</sup> Liquor Tank has been almost emptied liquor flow to the Digester is stopped and the remaining liquor in the Digester is drained down and pumped to the 2<sup>nd</sup> Liquor Tank through

the appropriate lines and valves. The Digester is now depressurized by opening the valves in the top line and the vapors passed to a Blow-Down Condenser, **35**. The alcohol-rich vapors are condensed and returned to the Recovery Alcohol Tank, **9**, for re-use in the process. The partially-delignified fibres are now sluiced from the Digester through the bottom valve, using water or preferably condensate from the evaporator. This sluiced pulp is sent to a tank, **51**, from which it is pumped continuously to conventional pulp refining, washing, screening, cleaning and bleaching operations. The liquor from these operations can be processed by conventional means for alcohol recovery and sodium acetate recovery.

The spent liquor under pressure in the Spent Liquor Tank, **4**, is flashed into a Flash Tank, **6**, and the vapors condensed through the Blow-Down Condenser, **35**, and returned to the Recovery Alcohol Tank, **9**, for re-use. The condensed liquor in the Flash Tank, containing the extracted lignin, is then pumped to the Lignin Precipitation Tank, **7**, where it is mixed rapidly with stillage from the Distillation Tower, **14**, and the pH adjusted to below 3.0 with acid and the mixture cooled to about 17°. Lignin precipitates from the mixture and forms a slurry. This is pumped to a suitable filtering device, such as a drum filter, **13**, where the lignin is removed as a wet cake that is sent to an appropriate drier, while the filtrate is pumped to a Recovery Feed Tank, **54**. From this tank the filtrate is pumped to an appropriately designed Distillation Tower, **14**. Such a tower would have a lower steam stripping section and an upper rectifying section, or be composed of two columns having these functions. In this tower alcohol, together with some esters, is recovered as an overhead condensate and returned to the Recovery Alcohol Tank for re-use in the process. Furfural, which is present in the filtrate, accumulates at one of the lower trays in the rectifier section where it is drawn off, cooled and mixed with water before being sent to the Decanter. The lower liquid phase in the decanter is crude furfural, which is upgraded to merchant furfural in a commercially available system. The upper layer is aqueous alcohol, which is passed back to the Distillation Tower to recover the alcohol by mixing with the tower feed stream. Steam to power the stream stripping section is provided by Reboilers at the bottom of the tower.

Aqueous stillage from the bottom of the Distillation Tower containing sugars, some lignin and minerals, is sent to an

appropriately designed multi-stage evaporator, **19**, where it is concentrated to about 25% solids. This concentrate is pumped to an Evaporator Concentrate Decanter, **23**, where a lower layer of oily lignin is recovered and dried. The upper aqueous layer containing xylose, xylans, other sugars and minerals, is sent to a commercially available xylose recovery unit, **24**, for production of purified xylose. The effluents from this unit include waste hexose sugars, which are passed to the fermentation operations for alcohol production, and an aqueous solution of minerals, which are returned to the cane fields as fertilizer.

The aqueous condensate from the Evaporator is passed to a commercially-available solvent extraction unit, **21**, such as those employing tri-octyl phosphine oxide, (TOPO), for recovery of acetic acid, formic acid, furfural and ethanol as separate marketable products. The clean water that exits this unit is useful in the pulp washing and bleaching operations.

Molasses is used as the fermentation raw material in a fermentation plant, **26**, employing yeast to produce ethanol and other fermentation products. This medium may be supplemented with additional sugars from acid-hydrolysis products of waste cellulose, as required for maximum productivity. Additional minerals that may be required at this stage can be supplied from the waste liquor stream of the xylose recovery plant. After fermentation is complete in a sequential battery of batch Fermenters, the beer is pumped to a Filter, **31**, where yeast and other solids are removed and dried for sale as high-protein animal feed supplement. The clarified beer is then passed to the Recovery Feed Tank, **54**, mixed with the filtrate from the Lignin Recovery Filter and pumped to the Distillation Tower for recovery of ethanol and other components.

By this invention a range of valuable products is produced from the solid and liquid residues of the cane sugar industry.